**Laboratory study of geological carbon sequestration using surrogate fluids:
Dielectric measurement and scaling of capillary pressure - saturation
relationships**

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Abstract

Multiphase flow models are powerful tools to understand and predict the flow and trapping of supercritical- CO_2 (scCO_2) in deep saline geologic formations. The constitutive relationship between capillary pressure (P_c) and saturation (S_w) is the essential input parameter into these multiphase models. By selecting surrogate fluids that match the density and viscosity ratios between scCO_2 and brine, the behavior of scCO_2 and brine may be reproduced under ambient conditions, making it possible to overcome the difficulty of conducting laboratory-scale experiments with scCO_2 . This paper presents the $P_c - S_w$ relationship for the surrogate fluids of scCO_2 and brine measured using the dielectric sensor method. To explore the necessity and applicability of measuring the entire $P_c - S_w$ relationship, we compared the dielectric sensor results with those from two scaling methods: classical Leverett scaling and a novel entry pressure scaling method. Additionally, the obtained surrogate fluid $P_c - S_w$ values were compared with experimental data of scCO_2 and brine under high pressure. The dielectric sensor method successfully obtained the $P_c - S_w$ relationship and produced more accurate measurements of residual and irreducible saturations compared to scaling methods.

Keywords: CO_2 sequestration; capillary pressure; surrogate fluids; dielectric sensor; scaling; interfacial tension; phase saturation

1. Introduction

Geological Carbon Sequestration (GCS) is considered a promising mid- to long-term large-scale atmospheric carbon mitigation strategy (IPCC, 2005). In implementing GCS, the flue gas of large industries is first captured to extract CO₂ (Benson and Surles, 2006; Elliot and Celia, 2012). This CO₂ is then injected and sequestered/stored in supercritical phase (scCO₂) in deep saline aquifers or depleted oil and gas reservoirs containing impermeable cap rocks that prevent its leakage (Juanes et al., 2006). The GCS scheme is being tested at various field sites (e.g., Torp and Gail, 2004; Chadwick et al., 2008; 2012; Wilson and Monea, 2004; Mathieson et al., 2010). Despite extensive studies on GCS, the mechanisms for effective scCO₂ storage still have many uncertainties that need to be understood (Schnaar and Diguilio, 2009). Many questions remain regarding the fundamental processes that lead to stable and efficient entrapment, particularly under naturally-occurring heterogeneous conditions. However, only limited insights on the contribution of heterogeneity can be obtained from field studies as the complex heterogeneity cannot be accurately characterized at all relevant scales. This lack of basic knowledge makes it difficult to predict the behavior and permanence of CO₂ injected into deep geologic formations.

Simulation models for predicting fluid flow require the relationship between capillary pressure (P_c) and saturation (S_w) (hereinafter, referred to as the $P_c - S_w$ relationship) as one of the essential input parameters. There are various approaches to obtain a $P_c - S_w$ relationship. A scaling method is a theoretical method for the determination of the $P_c - S_w$ relationship for a wetting and non-wetting fluid pair using a known relationship for another wetting and non-wetting fluid pair. Leverett scaling is a widely used method and uses the ratio of surface tensions of the two fluid pairs to scale the P_c for a reference fluid pair to that of the other pair (Leverett

1941). Experimental methods, on the other hand, to develop $P_c - S_w$ relationships for air-water are also well established (e.g., Schroth et al., 1995; Chen et al., 1999; Sakaki et al., 2008), and some of these methods have been extended to some non-aqueous phase liquid (NAPL)-water systems (Schroth et al., 1995; Chen et al. 1999).

Laboratory experiments have many advantages over field investigations, as the experimental conditions can be better controlled and the detailed behavior of injected CO_2 can be measured and/or visualized. Because of the high pressure/temperature required to keep CO_2 in the supercritical state, conducting experiments with scCO_2 is difficult or limited to simpler settings. However, larger-scale two or three-dimensional experiments are needed to study the effects of heterogeneity. The diffusive behavior of CO_2 can be monitored using X-ray and nuclear magnetic resonance imaging (Fleury et al., 2010; Krevor et al., 2012), but these instruments are not always easily available. Tokunaga et al. (2013) used unconsolidated sand in their laboratory experiments to capture capillary pressure data from a scCO_2 and brine system; however, it required a vessel which has a resistance to high pressure.

Use of surrogate fluids for scCO_2 and brine is advantageous because they can replicate the behavior of scCO_2 in a saline aquifer without the need to conduct experiments under high pressures or temperatures. Surrogate fluids have been used by Cinar et al. (2009), Pentland et al. (2010), Aminzadeh-goharrizi et al. (2012), and Trevisan et al. (2014) among others, as an analog to the scCO_2 – brine system. Cinar et al. (2009) chose fluids with viscosity, density, and interfacial tensions that are representative of scCO_2 when injected into formations with brine.

In our laboratory, a series of experiments were conducted in heterogeneous two-dimensional test tanks using surrogate fluids for scCO₂ and aquifer brine to investigate multiphase flow and capillary trapping mechanisms relevant to carbon sequestration (e.g., Trevisan et al., 2014). With the need to model these tank experiments in mind, we performed a direct measurement of the $P_c - S_w$ relationship for the surrogate fluid pair used in these tank experiments. In addition, the measured surrogate fluid $P_c - S_w$ values were compared with values obtained through two scaling methods as well as with measured $P_c - S_w$ values for scCO₂ systems previously obtained by Tokunaga et al., (2013).

2. Materials and Methods

In this study dielectric moisture sensors were applied to directly measure the $P_c - S_w$ relationship of the surrogate fluids (further described in Section 2.6). The efficiency and accuracy of the dielectric sensor method were then evaluated by comparing the measured $P_c - S_w$ with $P_c - S_w$ obtained using two scaling methods: Leverett scaling (described in Section 2.3) and our own developed method, which uses the ratio of entry pressures of the two fluid systems in the same porous medium (described in Section 2.4). Finally, to verify the applicability of the surrogate fluid $P_c - S_w$ measurements to CO₂ and brine systems, we compared our results to the CO₂/brine $P_c - S_w$ relationships of Tokunaga et al., (2013). Throughout this study, the $P_c - S_w$ experiments were conducted to measure both drainage and imbibition curves because hysteresis is known to have a significant effect on the capillary trapping mechanisms (Doughty, 2007; Juanes et al., 2010). Further explanation of the materials and test methods are provided below.

95

96 2.1 Test sands

97 Five uniform silica sands with different particle diameters (Accusands, Unimin
 98 Corporation, Minnesota, USA) were used in our experiments. These sands are identified by their
 99 sieve sizes; #12/20, #20/30, #30/40, #40/50, and #50/70, respectively. In addition, the sands have
 100 low organic content, high quartz content (99.8%), and the grain shape is classified as rounded
 101 (Schroth et al., 1995). Uniformity coefficients (d_{60}/d_{10}) are approximately 1.2 and the grain
 102 density is 2.66 g/cm^3 . Selected hydraulic properties of the test sands are given in **Table 1**. The
 103 van Genuchten parameters for air-water systems for each sand (Sakaki et al., 2013a) are also
 104 listed in **Table 1**. These parameters are required for the scaling methods, which derive the
 105 unknown $P_c - S_w$ relationship of a second pair of wetting and non-wetting fluids (i.e. the
 106 surrogate fluids) using the ratio of the known interfacial tension or entry pressures of the
 107 reference pair of fluids (i.e., water and air).

108

109 **Table 1.** Selected properties for the test sands measured for air-water system by Sakaki et al.
 110 (2013a).

Sand no.	Saturated hydraulic conductivity K_s (cm/s)	Intrinsic permeability k_i (cm^2)	d_{50} (mm)	S_r (-)	S_i (-)	Air Entry pressure head (cm of water)	van Genuchten parameters (drainage)		van Genuchten parameters (imbibition)	
							α (1/cm)	n (-)	α (1/cm)	n (-)
#12/20	0.38	3.8×10^{-6}	1.04	0.83	0.055	7.9	0.10	13.8	0.19	6.00
#20/30	0.24	2.3×10^{-6}	0.75	0.71	0.082	12.0	0.08	13.5	0.14	6.50
#30/40	0.11	1.1×10^{-6}	0.52	0.72	0.085	16.1	0.06	17.0	0.092	7.50

#40/50	0.052	5.3×10^{-7}	0.36	0.81	0.085	19.4	0.04	9.20	0.082	5.20
#50/70 ¹⁾	0.030	3.1×10^{-7}	0.27	0.80	0.093	34.0	0.03	17.0	0.050	5.50

¹⁾ Measured as part of this study using the Tempe cell apparatus. Abbreviation: S_r : residual water saturation (-); S_i : irreducible water saturation (-). The van Genuchten parameters were slightly modified from Sakaki et al. (2013a).

2.2 Selection of Surrogate Fluids for scCO₂ and Brine

Soltrol 220 (a paraffin oil, dyed with Sudan IV for visual observation, referred to here as Soltrol) and a glycerol/water mixture (80% glycerol by mass, referred to here as glycerol/water) were selected to replicate the dynamics of a system containing CO₂ and aquifer brine, respectively. The selection of the surrogate fluids was based on the comparison of the bond number, capillary number, viscosity ratio, and density ratio of the two fluid systems. These dimensionless numbers are defined as

$$\text{Viscosity ratio} = \frac{\mu_{nw}}{\mu_w}, \quad (5)$$

$$\text{Density ratio} = \frac{\rho_{nw}}{\rho_w}, \quad (6)$$

$$\text{Capillary Number} = \frac{\mu u}{\sigma}, \quad (7)$$

$$\text{Bond number} = \frac{\Delta \rho g k}{\sigma}, \quad (8)$$

where μ is the viscosity (M/L/T), subscripts _w and _{nw} refer to the wetting and non-wetting fluids, $\Delta \rho$ is difference in density between the two immiscible fluids (M/L³), g is the gravitational acceleration (L/T²), k is the intrinsic permeability (L²), u is the Darcy velocity (L/T), and σ is the interfacial tension (M/T²). These dimensionless numbers describe the

magnitude of the capillary force and dominant force (surface tension vs. gravity) applied to a fluid system. The dimensionless numbers for the Soltrol – glycerol/water pair are comparable to the scCO₂ – brine values (**Table 2**), indicating that a good replication of the flow behavior is likely. Potential effects of dissolution of scCO₂ into brine were neglected for simplicity. Furthermore, exsolution of CO₂ bubbles (e.g., Sakaki et al., 2013b; Plampin et al., 2014a, b) was not considered. This allowed us to consider only surrogate fluids that are immiscible with each other.

Table 2. Selected properties of scCO₂ and brine compared with the surrogate fluids. Ranges in scCO₂ and brine values are a result of differences between individual study salinities and temperatures.

	scCO ₂	Brine	Soltrol	glycerol/water
ρ (g/cm ³)	0.270 – 0.730 ¹⁾	0.95 – 1.23 ¹⁾	0.79 ²⁾	1.21 ⁴⁾
μ (mPa s)	0.023 – 0.061 ¹⁾	0.20 – 1.58 ¹⁾	3.90 ³⁾	60.9 ⁴⁾
μ_{nw}/μ_w (-)	0.03 – 0.20		0.064	
ρ_{nw}/ρ_w (-)	0.22 – 0.75		0.65	
σ (dyne/cm)	20.0 – 60.0		17.3 *	
Capillary number(-)**	10 ⁻⁵ – 10 ⁻⁸		10 ⁻⁶ – 10 ⁻⁷	
Bond number (-)**	10 ⁻⁷ – 10 ⁻⁸		10 ⁻⁵ – 10 ⁻⁶	
Dielectric constant	1.1-1.5 ⁵⁾	48-92 ⁶⁾	2.015 ⁷⁾	51.6-65.3 ⁸⁾
Solubility property	0-1.2 mol/kg ⁹⁾		Negligible	

¹⁾ Nordbotten et al. (2005), ²⁾ Kechavarzi et al. (2005), ³⁾ Fischer and Morrow (2006), ⁴⁾ the empirical formula from the study of Cheng (2008) was used to calculate density and viscosity of the Glycerol/water mixture at ambient conditions. ⁵⁾ Beckman (2004) ⁶⁾ Stogryn (1971), ⁷⁾ Capaccioli (2000), ⁸⁾ McDuffie Jr et al. (1962), ⁹⁾ Eke et al. (2011), * measured in this study. ** calculated using the sand properties in **Table 1**.

Dielectric properties of the fluids were also considered when selecting the surrogate fluids because a certain condition had to be met for the dielectric sensors to be used to measure the phase saturation of the surrogate fluids. Water and air have dielectric constants of 81 and 1, respectively (e.g., Topp et al., 1988; Chang et al., 2010). Quartz sand particles typically have a dielectric constant of about 4.7 (Robinson and Friedman, 2003). Because water has a dielectric constant value that is substantially higher than the values of air and sand particles, the apparent dielectric constant of a composite of the three phases strongly depends on the volumetric fraction of water. Soltrol has a dielectric constant of about 2 (Capaccioli, 2000). The dielectric constant of the glycerol and water mixture is between 51.6 and 65.3 (McDuffie Jr et al., 1962). Therefore, Soltrol and the glycerol/water mixture reasonably replicate this difference in dielectric constants. Under these conditions, the dielectric sensor showed sufficient sensitivity to the volumetric fraction of the glycerol/water phase for use as a saturation measurement.

2.3 Leverett Scaling Method

Leverett (1941) found that the $P_c - S_w$ relationship for one fluid pair in a porous medium could be calculated from the $P_c - S_w$ relationship of another fluid pair through the following dimensionless function, $J_{(S_w)}$:

$$J_{(S_w)} = \frac{P_{c,1}}{\sigma_1} \left(\frac{k_1}{\phi_1} \right)^{1/2} = \frac{P_{c,2}}{\sigma_2} \left(\frac{k_2}{\phi_2} \right)^{1/2}, \quad (1)$$

where P_c is capillary pressure (F/L^2), ϕ is porosity (dimensionless), and subscripts 1 and 2 refer to fluid pair 1 (reference fluid pair) and fluid pair 2 (scaled fluid pair). When scaling between

161 fluid pairs within the same porous medium (where the porosity and intrinsic permeability are the
162 same), P_c for fluid pair 2 can be calculated in terms of fluid pair 1 by

$$P_{c,1} = \frac{\sigma_1}{\sigma_2} P_{c,2}. \quad (2)$$

163 **Equation 2** shows that with Leverett scaling, the $P_c - S_w$ relationship for a fluid pair can be
164 estimated from the known $P_c - S_w$ relationship for a different fluid pair using the ratio of
165 interfacial tension values of the two fluid pairs.

166 A van Genuchten model curve (parameters in **Table 1**) was fit to values for P_c and S_w for
167 air and water (Sakaki et al., 2013a) to obtain a “reference” for scaling the $P_c - S_w$ curve for
168 Soltrol-glycerol/water according to **Equation 2**. Interfacial tension values for the air/water and
169 Soltrol-glycerol/water systems were measured using the ring method with a Cenco-DuNouy
170 tensiometer and DuNouy ring. It is based on measuring the force required to detach the ring from
171 the fluid surface (Fox and Chrisman, 1952). More details of the method can be found in Schroth
172 et al. (1995). A value of 73 dyne/cm was assumed for the air-water interface. Test periods for the
173 interfacial tension measurement were kept rather short (within 30 minutes) to avoid possible
174 contamination effects at the interface of the fluids. Soltrol with and without Sudan IV was used
175 to check whether mixing the dye changed the interfacial tension. All the experiments were
176 conducted at a room temperature of around 20 °C.

177

178 2.4 Entry Pressure Scaling Method

179 A primary limitation of the Leverett scaling method is that the scaling factor is solely
180 determined based on the interfacial tensions between the two fluids and it does not include the
181 possible effects of differences in the contact angle affecting the $P_c - S_w$ relationship for two fluid
182 pairs. Richardson (1961) incorporated contact angle into the Leverett scaling method, but contact
183 angle measurement between surrogate fluids such as Soltrol and glycerol/water mixture can be
184 problematic because bubbles tend to form on top of the glycerol/water mixture due to buoyancy.
185 As an alternative to a method that relies on the measurement of the contact angle, we propose
186 scaling based on entry pressure head, which would presumably account for the effects of both
187 contact angle and interfacial tension. To scale based on entry pressure head, we still use
188 **Equation 2**, except that we substitute the ratio of entry pressure heads in place of the ratio of
189 interfacial tensions. To convert the entry pressure head of Soltrol to the water head, **Equation 3**
190 was used (Mori, 2013).

$$h_d (\text{Soltrol, water head}) = h_d (\text{Soltrol, Soltrol head}) \times \frac{\rho_{\text{Soltrol}}}{\rho_{\text{water}}} \quad (3)$$

191 Scaling factors are then calculated by dividing the entry pressure head of the Soltrol in
192 terms of a water head by the entry pressure head of water as in **Equation 4** (Mori, 2013) to scale
193 the $P_c - S_w$ relationships of the air-water to the Soltrol-glycerol/water system.

$$scaling\ factor = \frac{h_d (Soltrol, \ water\ head)}{h_d (water, \ water\ head)} \quad (4)$$

To measure the entry pressure head of the Soltrol-glycerol/water system, the experimental set up shown in **Figure 1** was constructed. Three identical columns were placed in parallel on the same platform to verify the reproducibility of the results. The test sand was first tightly wet-packed (Sakaki and Illangasekare, 2007) in the glycerol/water mixture to a height of 15 cm. An external constant head device filled with glycerol/water, connected at the bottom of each column, was positioned at the same elevation as the surface of the glycerol/water-saturated sand (15 cm elevation) to maintain the glycerol/water head at the same level. A Mariotte bottle filled with Soltrol was connected to the top part of all three columns to supply Soltrol from above the glycerol/water saturated sand. The Soltrol injection pressure head was increased in a stepwise manner by elevating the Mariotte bottle at constant intervals of 0.5 cm (as shown in **Figure 1**). The entry pressure head was defined as the difference in elevation between the top of the sand and the top of the Soltrol within the column once a change in the outflow of the displaced glycerol/water was observed than converted into the height of water using equation 3. The Soltrol pressure head was measured 12 hours after raising the Mariotte bottle and photographed. When there was no longer glycerol/water outflow mass, the Mariotte bottle was raised by another 0.5 cm. This procedure was performed at room temperature (approximately 20 °C) for all five test sands listed in **Table 1**.

Figure 1. Experimental setup for the determination of entry pressure heads. Sand was saturated with glycerol/water in advance. The Mariotte bottle was raised to increase pressure, and drainage of glycerol/water was monitored at the drainage boundary fixed at the sand surface.

2.5 Dielectric Sensor Method

Both Leverett and entry pressure scaling methods assume that the irreducible (S_i) and residual (S_r) saturations are the same for the reference fluid pair and the scaled fluid pair; however, these values may vary among fluid pairs (Tokunaga et al. 2013), and the scaling methods are not able to capture these differences. The dielectric sensor method applied in this study allows for the direct measurement of all phase saturations, including S_i and S_r , during the generation of data for the $P_c - S_w$ relationships.

The column setup shown in **Figure 2** was used to directly measure $P_c - S_w$ relationships. The saturation of the glycerol/water mixture was measured using two EC-5 soil moisture sensors

(5.5 cm long, operation frequency 70 MHz, Decagon Devices Inc., Pullman WA, more details further described in Section 2.6) installed at 5 and 10 cm depths from the top of a 50-cm long vertically-oriented column. The sampling volume of the sensor is 18 cm^3 (Sakaki et al., 2008). Under the configuration shown in **Figure 2**, the sensors measure phase saturations that are averaged horizontally over the length of the electrodes ($\sim 5.5 \text{ cm}$).

Figure 2. The column setup for the dielectric sensor method. (Left) The initial condition of the experimental setup. (Right) After the drainage process has started, by lowering the constant head device.

The column experiment was performed for all five test sands listed in **Table 1**. The glycerol/water saturated sand was tightly wet-packed in the column as described in Sakaki and Illangasekare (2007). An external glycerol/water constant-head reservoir was connected to the bottom of the column to control the glycerol/water pressure. A Mariotte bottle filled with Soltrol

was connected to the top of the column. The elevations of the glycerol/water constant-head device and the Soltrol filled Mariotte bottle were initially set at the same level. The measuring process was similar to the process used for the entry pressure method described in Section 2.4. In this case, instead of increasing the height of the Mariotte bottle, the constant-head device was lowered in 2-4 cm height increments every 12 hours to increase the Soltrol pressure. **Figure 3** schematically illustrates the pressure distributions for the two fluids at equilibrium (Mori, 2013). This approach allows calculation of the capillary pressure in the surrogate fluids based on relative heights without directly measuring the capillary pressures of the two phases, using the following equation:

$$P_c = P_{nw} - P_w = \rho_{nw}gL - \rho_w gH, \quad (9)$$

where g refers to gravitational acceleration (L/T^2), and L and H correspond to heights (L) indicated in **Figure 3**.

Figure 3. Calculation of capillary pressure P_c based on hydrostatic pressure distributions in both fluids at equilibrium. Red line refers to non-wetting phase Soltrol and blue line is wetting phase glycerol/water.

As the drainage proceeded, Soltrol seeped further into the sand and the EC-5 sensor at the top started to register a decrease in sensor reading. The EC-5 sensor reading was converted into phase saturation S_w using the calibration. Calibration of the EC-5 sensor for Soltrol-glycerol/water was challenging, and it is described below. By changing the height of constant head device, the P_c and S_w values were obtained at each height and the $P_c - S_w$ relationship was constructed. When the glycerol/water constant head device was sufficiently low, and there were no detectable changes in S_w , it was assumed that S_w had reached the residual saturation, S_r . The wetting cycle was then initiated by elevating the glycerol/water reservoir in a stepwise manner.

2.6. Calibration of Dielectric Sensor

The EC-5 soil moisture sensors were used to monitor the glycerol/water saturation measure capacitance of the surrounding soil. The surrogate fluids were selected so that the measured capacitance is a function of the volumetric fraction of Soltrol, because it has a dielectric constant appreciably less than that of the glycerol/water (a ratio of approximately 2 to 51.6-65.3 as indicated in Section 2.2).

Calibration of the EC-5 sensors was based on the two-point mixing model (**Equation 10**) developed for an air-water system (Sakaki et al., 2008; 2011).

$$S_w = \frac{X^N - X_{Soltrol}^N}{X_{Glycerol}^N - X_{Soltrol}^N} \quad (10)$$

where X is the observed sensor reading, and $X_{Soltrol}$ and $X_{Glycerol}$ are reference sensor readings under fully saturated conditions for Soltrol and the glycerol/water mixture, and N is a fitting parameter, respectively.

Sensor readings were taken in several saturated soil samples containing varied percentages of each liquid phase (Soltrol and glycerol/water) (**Figure 4, left**). Unlike in an air-water system, we were unable to mix the two surrogate fluids in sand without entrapping air because the lighter Soltrol would quickly flow upward in the form of bubbles and ganglia. We found that the EC-5 sensor output was sensitive to the vertical Soltrol and glycerol/water separation within the sand. However, by emplacing the sensor vertically through the sample (such that it crossed the Soltrol and glycerol/water boundary within the sample), we could measure values with the sensor that were sensitive to the phase percentages and were analogous to values that would be measured in a well-mixed samples.

The sensor reading (X) was recorded and the mixing model (**Equation 10**) was fitted to the measured sensor reading data. The exponent N was determined for the Soltrol-glycerol/water system. Note that N is specific to sensor type (Sakaki et al., 2008; 2013a) and possibly to fluid pairs. The best-fit value of N was found to be 2.5, which is identical to that obtained by Sakaki et al. (2008; 2011) for the air-water system. The measured calibration data points and the fitted mixing model are shown in **Figure 4 (right)**.

Figure 4. Calibration of the EC-5 sensor. (Left) An example calibration sample. (Right) Determination of fitting parameter in the mixing model. The symbols are measured raw sensor outputs at different saturations and the line was obtained by fitting the mixing model (**Equation 10** with $N = 2.5$).

3. Results and Discussion

3.1 Leverett Scaling

The average value of the three repeated interfacial tension measurements is summarized in **Table 3**. Adding Sudan IV to Soltrol did not significantly affect the interfacial tension. The measured interfacial tension was 17.3 with Sudan IV and 16.7 dyne/cm without it. The Leverett scaling factor was calculated to be 0.24 from the interfacial tension values of air-water (73 dyne/cm) and Soltrol-glycerol/water (17.3 dyne/cm). **Figure 5** shows the predicted $P_c - S_w$

relationships for the Soltrol-glycerol/water system for each sand size based on Leverett scaling of the air-water system (**Equation 2**) as well as the original $P_c - S_w$ relationships for the air-water system.

Table 3. Interfacial Tensions of different combination of fluids and the Leverett scaling factors.

Fluid combination	σ [dyne/cm]	Temperature [°C]	$\frac{\sigma}{\sigma_{air/water}}$
<i>Overview of literature ¹⁾</i>			
Soltrol (without sudanIV)-water	36.4	20.0	0.50
<i>Independently measured by Cenco-DuNoüy tensiometer *</i>			
Air-water	73.0	17.6	1.00
Glycerol/water-Soltrol (without sudanIV)	16.7	17.7	0.23
Glycerol/water –Soltrol	17.3	19.0	0.24

¹⁾ Schroth et al. (1995), * mean of three measurements, the measured values were almost identical

Figure 5. Scaled $P_c - S_w$ relationships for the surrogate fluids in five Accusands using Leverett scaling factor (red). Unscaled reference $P_c - S_w$ relationships for air-water are also shown for comparison. Note that S_i and S_r are assumed to remain unchanged.

314

315 3.2 Entry Pressure Scaling

316 Entry pressure heads (h_d) for Soltrol added to sand saturated with the glycerol/water
 317 mixture were obtained for each of the test sands. These values are compared to the air-water
 318 system pressure heads of Sakaki et al. (2013a) in **Table 4**. The scaling factors obtained for each
 319 of the five Accusand samples were in the range of 0.24-0.38. The entry-pressure scaled $P_c - S_w$
 320 relationships for each sand size are plotted in **Figure 6**. Each sand sample had somewhat higher
 321 scaling factors using entry-pressure scaling than factors calculated through Leverett scaling,
 322 except for the #12/20 and #50/70 sands, which yielded similar values using both scaling methods.
 323 The results from the entry pressure method are likely more representative of the actual $P_c - S_w$
 324 relationship than those inferred from Leverett scaling because the entry pressure method includes
 325 the effects of contact angle while Leverett scaling does not.

326

327 **Table 4.** Measured entry pressure and entry-pressure scaling factors

Sand no.	Measured h_d of Soltrol-glycerol/water system (cm of water)			h_d of visual entry at wall (cm of water)	Average h_d of glycerol-Soltrol system (cm of water)	h_d of air-water system (cm of water)*	Scaling factor (-)
	Column 1	Column 2	Column 3				
#12/20	1.9	1.9	1.9	0.8	1.9	7.9	0.24
#20/30	3.9	4.0	4.1	1.9	4.0	12.0	0.33
#30/40	5.2	5.2	5.2	4.4	5.2	16.1	0.32
#40/50	7.3	7.5	7.0	4.8	7.3	19.4	0.38
#50/70	11.1	-	11.1	4.0-4.8	11.1	34.0	0.24

328 *Values taken from Sakaki et al. (2013a)

Figure 6. Scaled $P_c - S_w$ relationships for the surrogate fluids in five Accusands using entry pressure scaling (green). Unscaled reference $P_c - S_w$ relationships for air-water are also shown for comparison. Note that S_i and S_r are assumed to remain unchanged.

Visual observations showed that the red-dyed Soltrol started to penetrate the sand pack somewhat before a significant outflow of glycerol/water was observed. When this “visual” entry was seen, only small outflows of glycerol/water were confirmed. This was then followed by a more distinct outflow (as well as an inflow of Soltrol), after the Soltrol pressure increased. Therefore, the entry pressure values determined based on the significant mass inflow/outflow were larger than those visually determined (values given in columns 4 and 5 of **Table 4**).

It has to be emphasized here that visual observations of the column wall only present what is happening at the interface of column wall and sand. Unless other methods such as light transmission are used, no information on what is occurring “in” the sand column is available from the images. It is well known that at the column-grain interface, the grains are forced to align along the column wall and the grain pattern is no longer random. This leads to the porosity along the column wall being locally higher (i.e., wall effects). This thin layer along the column wall with a larger porosity (or equivalently larger pore) allows glycerol/water to drain and Soltrol to enter at a lower entry pressure. A similar observation was reported by Culligan et al. (2006). We believe that this is what is seen in the photographs, which led to apparently smaller entry pressure values. **Figure 7** shows photographs after Soltrol penetration along the wall was observed. This artifact can be a pitfall if the entry pressure measurement relies only on visual observation. Therefore, it is highly recommended to measure entry pressure based on flow mass rather than using visual observation.

Figure 7. Soltrol penetration along the column wall-sand grain interface at pressures below the entry pressure. No penetration was observed in the bulk of the sand even when Soltrol had penetrated along the interface.

3.3 Dielectric sensor results

Measured $P_c - S_w$ values for each sand sample were plotted and fit with the van Genuchten model (**Figure 8**). The fitted van Genuchten parameters are summarized in **Table 5**. The S_i and S_r observed in the measurements (also shown in **Table 5**) with Soltrol-glycerol/water were used to estimate the fitting parameters. Because of the uniform grain size distribution of the test sands used in this study, a rapid change in S_w with a small change in P_c was observed, leading to flat $P_c - S_w$ curves. Although the number of data points for the coarsest #12/20 sand was somewhat limited, measurements were repeated twice for each test sand, and the results showed reasonable repeatability. The saturation values measured with the dielectric sensors were unaffected by the wall effect and consequently accurately reflected the actual drainage/wetting

behavior in the test sands. The uncertainty in the estimated saturation resulting from the scatter in the calibration data (**Figure 4**) was estimated to be $\pm 0.03 \sim 0.07$.

Table 5. van Genuchten model parameters fitted to the $P_c - S_w$ data for the Soltrol-glycerol/water system obtained with the dielectric sensors. Note that S_r and S_i are the actual measured values for Soltrol-glycerol/water.

Sand no.	<i>Measured</i> S_r (-)	<i>Measured</i> S_i (-)	Soltrol Entry pressure in water head (cm)	van Genuchten parameters (drainage)		van Genuchten parameters (imbibition)	
				α (1/cm)	n (-)	α (1/cm)	n (-)
#12/20	0.857	0.104	1.9	0.29	20.24	0.85	15.54
#20/30	0.734	0.056	4.0	0.29	6.61	2.03	2.45
#30/40	0.739	0.076	5.2	0.21	6.56	0.60	4.59
#40/50	0.836	0.040	7.3	0.17	4.47	0.36	3.50
#50/70	0.703	0.088	8.0	0.10	5.05	0.54	3.03

Figure 8. Measured $P_c - S_w$ relationships for the surrogate fluids in five Accusands using dielectric sensor method (open and closed triangles). Unscaled reference $P_c - S_w$ relationships for air-water are shown for comparison. Also shown in blue are the best-fit van Genuchten models.

3.4 Comparison of the Three Methods

Although Leverett scaling is the easiest method of the three tested here, higher capillary pressures were calculated using Leverett scaling than with entry pressure scaling or the dielectric sensor method, which both rely more on physical data than Leverett scaling (**Figure 9**). Significant differences in entry pressure did not result in large changes in interfacial tension (**Table 6**). Tokunaga et al. (2013) measured scCO₂ – brine interfacial tensions under two different pressures and showed that Leverett scaling was insufficient to scale the capillary pressure for different reservoir conditions.

Table 6. Comparison of fluid dependent systems

	#40/50	#50/70	Tokunaga et al. (2013) at 8.5 MPa	Tokunaga et al. (2013) at 12.0 MPa
Grain size (mm)	0.360 ¹⁾	0.270 ²⁾	0.250-0.355	0.250-0.355
Porosity (-)	0.34 ¹⁾	0.34 ²⁾	0.381	0.381
μ_{nw}/μ_w (-)	0.064	0.064	0.033 ³⁾	0.075 ³⁾
ρ_{nw}/ρ_w (-)	0.65	0.65	0.27	0.636
σ (dyne/cm)	17.3	17.3	33.1	30.1
Capillary Number (-)	$10^{-6} - 10^{-7}$	$10^{-6} - 10^{-7}$	$10^{-8} - 10^{-9}$	$10^{-8} - 10^{-9}$
Bond Number (-)	$10^{-5} - 10^{-6}$	$10^{-5} - 10^{-6}$	10^{-5}	10^{-5}
Entry pressure (kPa)	0.712*	0.787*	0.8**	0.5**
S_i	0.040	0.088	0.04-0.05	0.03-0.05
S_r	0.812	0.703	0.92-0.93	0.66-0.81

¹⁾Sakaki et al. (2013a), ²⁾measured as part of this study, ³⁾El-Dessouky and Ettouney (2002),

*surrogate fluids, **scCO₂-brine, Tokunaga et al. (2013) sieved 40-60 mesh Unimin round

quartz sand to obtain the grain size range listed above. Their sand hydraulic conductivity was 3.9×10^{-4} m/s.

All the methods captured the hysteretic behavior well. In most of the sands, the scaling methods resulted in higher calculated capillary pressures during imbibition than were calculated using the dielectric sensor method. This is consistent with the $P_c - S_w$ relationship measured for scCO₂ and brine under high pressure by Tokunaga et al. (2013). This suggests that the scaling factors may need to be determined separately for drainage and imbibition cycles; this is perhaps because the interfacial tensions are different at advancing and retreating fronts.

For some of the test sands, S_r was poorly defined due to the shape of the $P_c - S_w$ data; in those cases, we carefully selected S_r by choosing a value where the capillary pressure is slightly below zero. However, there were distinct differences in both S_i and S_r compared with those for the air-water system (**Table 7**). These differences between the air-water and glycerol/water values highlight the advantage of using the dielectric sensor method over the scaling methods. There was no clear trend in the differences among S_i and S_r with respect to the different sands.

Table 7. Comparison of S_i and S_r values between air-water and Soltrol-glycerol/water systems

Sand no.		air-water ¹⁾	Soltrol-glycerol/water
#12/20	S_i	0.055	0.104
	S_r	0.827	0.857
#20/30	S_i	0.082	0.056
	S_r	0.712	0.734
#30/40	S_i	0.085	0.076

#40/50	S_r	0.725	0.739
	S_i	0.085	0.040
	S_r	0.812	0.836
#50/70	S_i	0.093	0.088
	S_r	0.703	0.703

411 ¹⁾ Values taken from Sakaki et al. (2013a)

Figure 9. Comparison of $P_c - S_w$ curves for the surrogate fluid pair obtained using the Leverett scaling method, the entry pressure scaling method, and the dielectric sensor method.

417

418 3.5 Comparison with scCO₂/brine data

419 Although Tokunaga et al. (2013) measured scCO₂-brine capillary pressure in
420 unconsolidated sand under two different high pressure/temperature test conditions, there were
421 some similarities between the observed $P_c - S_w$ curves for the #40/50 and #50/70 Accusands used
422 in this study and the $P_c - S_w$ curves for Tokunaga et al. (2013) (**Figure 10**). Low S_i values were
423 observed in both scCO₂-brine and Soltrol-glycerol/water, the Soltrol-glycerol/water fluid
424 properties were relatively close to those in the Tokunaga et al. (2013) 12 MPa measurements
425 (**Table 7**), and the S_i and S_r values for Accusand #40/50 were similar to those in their 12 MPa
426 system. However, the entry pressure in their 12 MPa was distinctly different from our results,
427 and this could be explained from the wettability changes with exposure time because high
428 wettability lowers the entry pressure value.

429 Nonetheless, the surrogate fluids and actual scCO₂ and brine displayed qualitatively
430 similar capillary pressure behavior and reasonable quantitative agreement with allowances for
431 differences in the sand properties. This similarity in $P_c - S_w$ behavior between the surrogate
432 fluids and scCO₂/brine demonstrates that surrogate fluids are applicable to studies of scCO₂/brine
433 system behavior.

Figure 10 Comparison of $P_c - S_w$ relationships by Tokunaga et al. (2013) with those for Accusand #40/50 and #50/70; (a) drainage cycle, and (b) imbibition cycle.

4. Conclusions

We evaluated the dielectric sensor method to obtain the $P_c - S_w$ relationships for the surrogate fluids and compared the results with those from two scaling methods as well as literature data measured for scCO₂/brine. As the surrogate fluids, we selected Soltrol and glycerol/water mixture, which had a similar viscosity, density ratio, and interfacial tension to those of scCO₂ and brine at typical sequestration conditions. In addition, the fluids were selected because they had a distinct difference in their dielectric constants, which allowed for the dielectric sensors to detect the phase saturation of the system. The following statements summarize characteristics of each method.

- 1) Leverett scaling is perhaps the simplest and quickest method to obtain a roughly estimated scaling factor for the selected surrogate fluids. The method, however, requires a device to measure interfacial tension for both fluid pairs. Furthermore, our data suggest that the scaling factor may be different for the drainage and imbibition cycles.
- 2) The entry pressure scaling method involves a relatively simple process that can be performed without any special devices. The disadvantage of this method was, like Leverett scaling, the scaling factor may need to be obtained separately for the drainage and imbibition cycles. Also noteworthy is that the entry pressure should be based on mass flow and not on visual observations such as photographic images of the column wall. An advantage of the entry pressure scaling method over Leverett scaling is that entry pressure scaling incorporates effects of both contact angle and interfacial tension.
- 3) The dielectric sensor method is more time-consuming but the direct measurement of $P_c - S_w$ data yielded S_r and S_i . For the surrogate fluid pair that we used, S_r values were not far from those of the air-water system. However, the method required some knowledge on calibrating the dielectric moisture sensor for this special pair of fluids. Furthermore, this method may not work as well for fluid pairs with a small contrast in the dielectric properties.
- 4) The dielectric sensor method yielded reasonable measurements of the $P_c - S_w$ relationship for the surrogate fluids compared to the scaling methods, although neither scaling method agreed completely with the dielectric sensor data. At high saturation, the entry pressure scaling method agreed more closely with the dielectric sensor method, whereas at lower saturation Leverett scaling yielded a closer fit with the dielectric sensor data. The deviation between the dielectric sensor method and two scaling methods was lower during drainage than during

imbibition. Although it involved some extra setup, the dielectric sensor method provided the most direct, complete, and accurate measurement of the capillary pressure relationships.

5) Although capillary pressure measurements were influenced by differences in the sand and fluid properties, our data with surrogate fluids seemed to be consistent with the scCO₂-brine data of Tokunaga et al. (2013). We therefore, find that our surrogate fluids approach under low pressure environments can be used to gain insight into scCO₂-brine behavior at reservoir conditions. The ability to study this behavior under low pressure conditions will make it much more feasible and practical to study larger-scale heterogeneous systems in a controlled environment.

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